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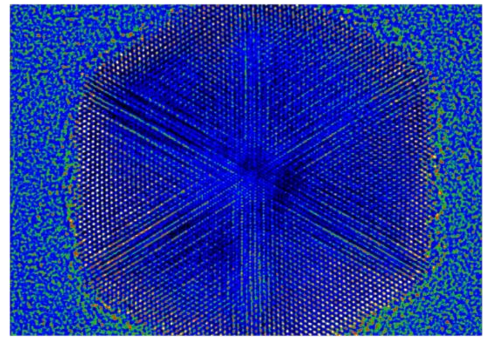
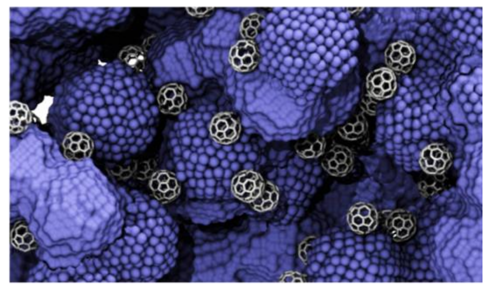
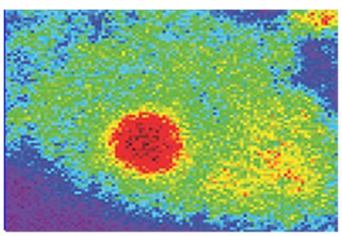
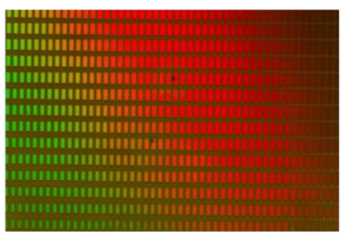
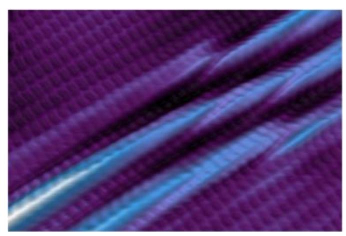
CENTER FOR NANOSCALE MATERIALS

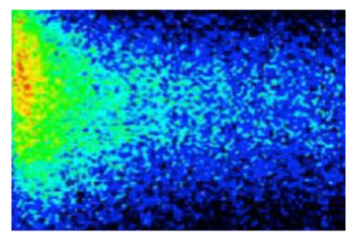












STRATEGIC PLAN 2016-2020

Argonne National Laboratory
Nanoscience and Technology Division
U.S. Department of Energy, Office of Science,
Office of Basic Energy Sciences

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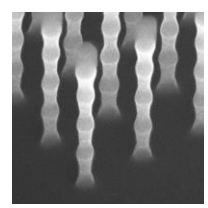
Executive Summary

The Center for Nanoscale Materials (CNM) at Argonne National Laboratory is a premier user facility providing expertise, instrumentation, and infrastructure for interdisciplinary nanoscience and nanotechnology research. As a Department of Energy funded research center, the CNM is at the forefront of discovery research that addresses national grand challenges encompassing the topics of energy, materials and the environment. Under the overarching scientific theme of ***Manipulating Nanoscale Interactions for Energy Efficient Processes****,* we seek to discover new materials, visualize events with high resolution as they occur, understand the physics and chemistry of energetic processes at the nanoscale, and manipulate nanoscale interactions to synthesize and fabricate useful, energy efficient structures with new functionalities**.** Much of the focus of the past decade of work in nanomaterials has been on understanding the properties of materials at the nanoscale, and developing ways of synthesizing and manipulating them. The tasks for the next decade will include the hierarchical integration of materials across different length scales (from the nano to the meso) in order to create energy efficient and affordable functionality that affects the public good in a major way. They will also include manipulating and extracting detailed information at the extremes of temporal, spatial and energy resolutions, down to single atom and molecular entities.

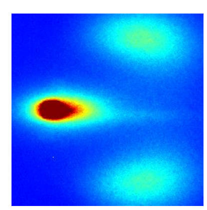
Our strategy is closely linked to Argonne's strategic plan that guides core research programs at Argonne, and focuses on discovery science and engineering while complementing other DOE user facilities at Argonne, namely the Advanced Photon Source (APS) and the Argonne Leadership Computing Facility (ALCF). We interact with these facilities where appropriate, thereby allowing us to leverage developments across the lab to enhance the world-leading capabilities available to the CNM users and make available to them a wider range of tools and expertise.

The CNM mission for user support and staff science is executed through six research groups. Cross-cutting activities, which link the six research groups, result in multiple joint projects. The scientific thrusts of the six research groups are described below:

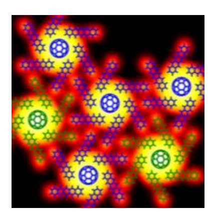
- **The Nanofabrication & Devices Group** (NFD) studies the fundamental science behind the development of micro- and nanoscale systems with the goal of achieving unprecedented control in the fabrication, integration, and manipulation of nanostructures. This includes the incorporation – under cleanroom conditions – of materials and active submicron elements that couple mechanical, optical, and electrical signals to produce working nanofabricated structures.



- **The X-ray Microscopy Group** (XMG) uses high resolution x-ray imaging to visualize and quantify novel nanoscale electronic, magnetic, and phonon phenomena in materials, particularly in embedded structures and systems under operando conditions. The group harnesses the high brightness of the APS, nanofocusing x-ray optics and imaging methods to provide quantitative two and three-dimensional insight into nanoscale structure and interactions. Our research exploits frontier concepts for coherent and scanned probe imaging for study of both ordered and disordered matter, from the scale of devices down to single defects and atoms.

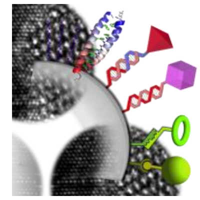


- **The Quantum & Energy Materials Group** (QEM) designs and studies atomic-scale to meso-scale materials with implications for energy, the environment, and coherent information transfer/sensing. Their research includes (i), using a powerful suite of scanning tunneling probe characterization and atomic/molecular manipulation capabilities to "design-in" engineered quantum states down to single atoms,

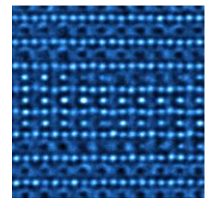


molecules or defects; and (ii), control molecular and nanoscale interactions for the accelerated discovery and fundamental understanding of artificial three- and two-dimensional materials for energy, reaction chemistry, and pollution remediation.

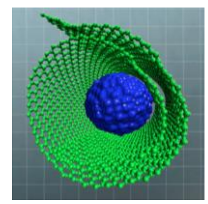
- **The Nanophotonics & Biofunctional Structures Group** (nPBS) studies optical processes at the extremes of time and space resolution through ultra-fast spectroscopies and advanced microscopies. The goal is to understand and realize efficient energy transduction in nanostructures, and study the physics of coherent radiative processes for quantum sensing. The group also seeks to create novel biological assemblies for nature-inspired studies of energy conversion, coherent energy transport, and biosensing mechanisms in cell-like environments functionalized with engineered nanomaterials. The group combines the properties of metals, organics, semiconductors and dielectrics to synthesize efficient catalysts, chemical and biological sensors, and hybrid biological moieties.



- **The Electron Microscopy Center** (EMC) develops new research capabilities that go beyond off-the-shelf technology in conjunction with the broadest scientific community that helps identify, define, and develop transmission and analytical electron microscopy needs to address the science of the future. Current work focuses toward the development of a large-gap lens configuration to accommodate enhanced multimodal capabilities and upon which fast sources and detectors can be incorporated.



- **The Theory & Modeling Group** (TMG) works on large scale molecular dynamics, high-level electronic structure theory, quantum and electrodynamics, multi-scale modeling and data science based approaches to understand and predict a wide range of phenomena including nanoscale tribology, thermal and charge transport, and quantum entanglement in hybrid plasmonic systems.



The CNM provides world-leading expertise and tools to its users, and some of the unique capabilities available at CNM include the Hard X-ray Nanoprobe, novel scanning tunneling microscopes, ultrafast optical spectroscopy techniques and single photon emitters, nanomechanical and plasmonic structure fabrication, chromatic aberration corrected TEM, and the Carbon high-performance computing cluster. The CNM currently employs 75 staff, who contribute to world-leading scientific programs in addition to supporting the users of the facility. During FY15, the CNM hosted 529 users from academia, national laboratories and industry. CNM users, staff, and post-docs are engaged in high-quality science, as evidenced by the publication of a total of 739 papers during the past three years with 31% of the papers in the top 20 highest impact journals as defined by DOE BES, and by numerous invitations to present their work at top conferences worldwide.

The CNM considers its role to be one that has both an anticipatory component, where we are ready to provide capabilities users will need 2-5 years into the future, as well as a steering component where our scientists help influence and shape the direction where nanoscience (and users) should be headed. Four specific areas where we will increase our activity for the future are: energy efficient architectures, nonlinear photonics and nanomechanics, quantum materials and sensing, and the confluence of first principles physics and data science for nanomaterials research. Straddling these areas of impact will be our cross-cutting characterization activities in three dimensional x-ray imaging that harness the high brightness of the Advanced Photon Source, and electron microscopy techniques that push towards lab-in-a-gap concepts. Our future staffing and “new capability” capital equipment purchases will reflect these directions. These strategic areas span all six research groups at the CNM. The accompanying documents describe the cross-cutting research efforts of the groups, as well as our over-arching strategy and its planned execution.

1. Introduction

In order to maintain a position at the forefront of nanoscience and nanotechnology, and to maintain excellence as a world-class user facility, the CNM will continue to pursue innovative research programs and develop new capabilities. Strong synergy and cross-cutting activities link the six research groups within the CNM, leading to a whole that is greater than the sum of the parts. Since its beginning the CNM has cultivated an engaged and productive user community, becoming a resource used by a wide spectrum of researchers from across the U.S., as well as from Europe, South America, and Asia. During FY2015 there were 529 users who took advantage of the leading-edge capabilities offered by the CNM. As evidenced by recent survey comments, CNM staff members have gone beyond simply providing excellent research facilities to contributing integral aspects of the resulting scientific outcomes. Some recent user comments include:

> *"The staff members at the facility are very knowledgeable. They are not only familiar with their instruments but also with their users' research. They are very dedicated in the user support including experimental design, training and technical assistance."*
>
> *"All workers at CNM have been extremely helpful on all of our visits. Can't say enough great things about them."*
>
> *"All the staff are excellent. I have enjoyed working on projects with them over the years. We appreciate access to CNM facilities and strong support of CNM staff."*

***Figure 1-1**. Integrated approach to research at the CNM reflecting close links between the CNM user program, staff expertise, and state-of-the-art facilities for nanoscience and nanotechnology research.*

Figure 1-2. *Center for Nanoscale Materials staff members in 2016.*

Within the user facility framework, the CNM has a unique opportunity for growth and interactions with universities, scientific organizations and research institutions from across the globe. We offer access to diverse scientific expertise and an interdisciplinary research culture. Examples include experienced staff in nanofabrication capable of tackling challenging issues in materials and nanostructural design; recent success stories in this area include hybrid materials with superlubricity properties, MEMS devices for manipulating x-rays, and metasurfaces for controlling optical wavefronts. Our expertise in nanophotonics has enabled probing ultrafast optical responses of photoexcited nanostructures, demonstration of a bio-inspired nanoscale hydrogen generator, and development of techniques for the ultrasensitive ratiometric detection of toxins. Our expertise in crystal growth and electron microscopy has enabled visualization and determination of the mechanisms of hybrid nanoparticle growth.

Our Electron Microscopy Center utilizes a chromatic aberration corrected TEM and has recently shown it possible to distinguish between atoms such as Ca and Ba in the sublattices of complex oxides. The chromatic aberration corrected microscope, one of only three worldwide offers the particular benefit of being able to probe thicker samples (compared to other microscopes) without loss of resolution. The design of novel materials has been guided by the development of integrated computational tools with predictive capabilities. These studies have contributed to the search for low cost, earth abundant alternatives to platinum for applications as catalysts, and the explanation of unique superlubricity effects observed in nanoscale graphene-nanodiamond lubricants.

The scanning probe microscopy techniques (SPM) developed in CNM have been utilized to discover and characterize new materials. A recent example is our recent discovery of a new 2D material, borophene, with unexpected metallic character. Our Hard X-ray Nanoprobe (HXN) is the only dedicated x-ray microscopy facility within the Dept. of Energy's Nanoscale Science Research Centers. Recently the unique insights gained by three dimensional high resolution (~30 nm) imaging using HXN was critical to discovering a new intermediate phase in lead-free ferroelectric materials, as well as significant structural improvements in SiGe nanomembrane-based materials for semiconductor quantum electronics. Our expertise in SPMs also enabled the development of the world's first dedicated synchrotron beamline for SX-STM. When completed, this SX-STM user facility will combine two powerful capabilities: enabling orbital level imaging of molecules

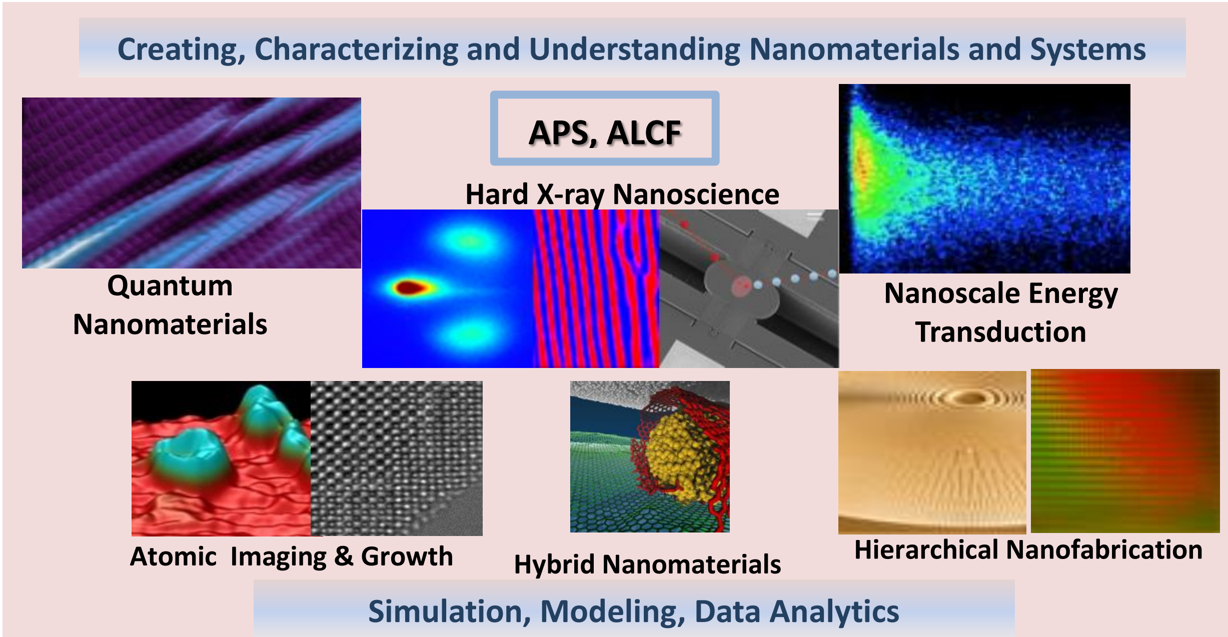


Figure 1-3. *Illustration of CNM's unique capabilities among the five DOE premier user centers for interdisciplinary research at the nanoscale. Overarching (top) and crosscutting (bottom) research is shown as blue highlighted banners. The Advanced Photon Source (APS) and Argonne Leadership Computing Facility (ALCF) are Argonne's other major BES facilities.*

and nanostructures, and being able to chemically identify atoms. These imaging capabilities are now being extended to rapidly visualize elemental and chemical processes in biological systems with unprecedented resolution.

Biology remains an inspiration for, and the model for, complex, hierarchical, functional material architectures that operate at high energy and/or quantum efficiency. Natural systems adopt a large degree of inhomogeneity and disorder to evolve and achieve resilience by harnessing environmental fluctuations. Using biology as well as the inorganic world for inspiration, our ultimate goal is to: (i), understand the physics and chemistry of energy transfer mechanisms at the extreme limits of spatial and temporal resolution, and (ii), create artificial energy efficient systems that can adapt to, evolve, and self heal as they are exposed to their operating conditions. Consequently, the CNM has identified *Manipulation of Nansocale Interactions for Energy Efficient Systems* as a guiding principle for our strategic scientific vision.

2. Vision for Scientific Growth

Understanding how the properties of known materials change when scaled down to the nanometer regime, and developing ways of synthesizing and manipulating materials at the nanoscale, dominated the past decade of nanoscience. The success of nanoscience for the future, however, will depend on understanding how to manipulate the hierarchical integration of multifunctional materials across different length scales to create advanced and affordable functionality. Future progress in the nanosciences will depend upon a multidisciplinary approach to synthesizing, connecting, measuring and embedding nanomaterials within usable architectures, with the emphasis on a system level rather than individual process level view. The nanosciences have also, so far, been very much a play between physics, chemistry, materials science and the biological sciences. We see a future vision for the field that will include extensive interaction with computer scientists, architects and data scientists. A schematic of the progress of materials science, followed by the emergence of the nanosciences is shown in Figure 2-1.

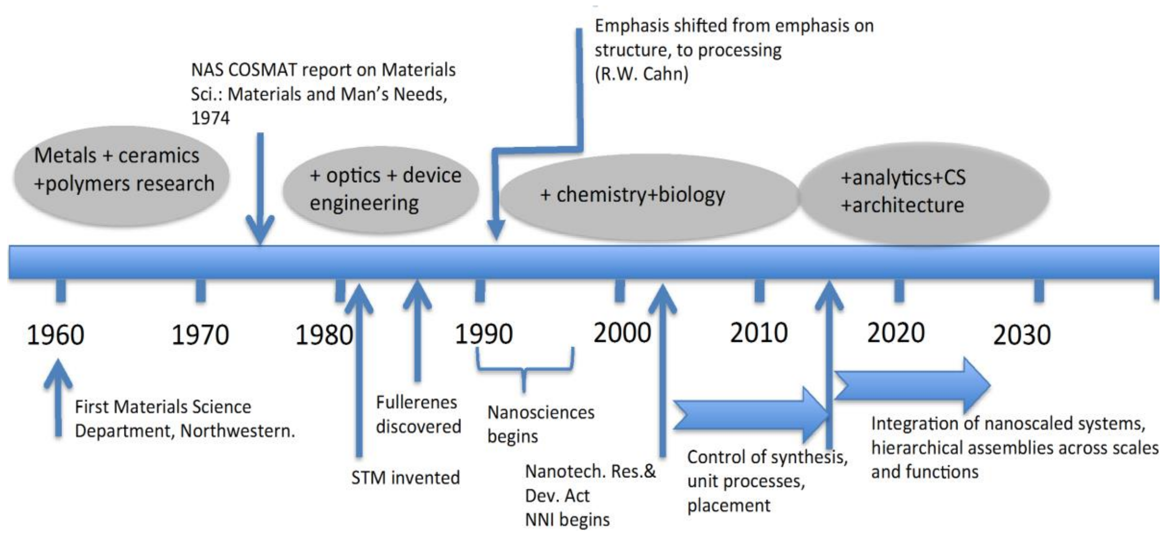


Figure 2-1*. Schematic showing the emergence of materials science as a formal discipline, followed by the emergence of the nanosciences and its trajectory.*

A recent Basic Energy Sciences Advisory Committee (BESAC) report from the DOE Office of Basic Energy Science (BES) on *Challenges at the Frontiers of Matter and Energy: Transformative Opportunities for Discovery Science* highlights the necessity for developing approaches for energy efficient systems by creating hierarchical assemblies, in particular those that involve realistic non equilibrium structures. A recently concluded workshop by the BES *Basic Research Needs for Quantum Materials for Energy Relevant Technology* focused on the emergence of quantum materials, and for exploring the physics of individual quantum states and entangled states for coherent information processing with minimal loss in energy. This has been driven by the needs of quantum information processing, developing sensors that go below classical noise limits, and is timely due to the emergence of new capabilities for manipulation and controlling materials at the nanoscale and techniques for computing their properties. There is tremendous opportunity in exploiting new materials with controlled and deterministically placed defects and heterogeneities for creating quantum states that would form the basis for such quantum information systems. Similarly, a workshop titled *Neuromorphic Computing: From Materials to Systems Architecture* co-organized by BES and the Advanced Scientific Computing Research (ASCR) Offices of DOE highlighted the influence of neuromorphic systems and biological information processing for ultra-low power computing for the future. This translates to an interest in building materials systems and controlling defects in materials that can be controllably altered by low energies (and low voltages of ~100s of mV), and where inhomogeneity and disorder is embodied in architectures that embrace variability. The recently explosion of data analytics, the availability of cheap computing for massive scale statistical computing and the OSTP initiated thrust on materials genomics has created significant interest in how machine learning combined with first principles physics can influence computational materials science. Finally, the proximity of the Advanced Photon Source (APS) and the anticipated 100 times improvement in coherent x-ray intensity in the near future call for new techniques that that expand the study of soft matter, of reaction chemistry, and time resolved studies. A marker for future user interest and research involvement may be found in the Grand Challenges identified by DOE, NSF and other government organizations shown in Figure 2-2. Our research vision is in full alignment with these challenges, keeping us ready for user interactions in the future.

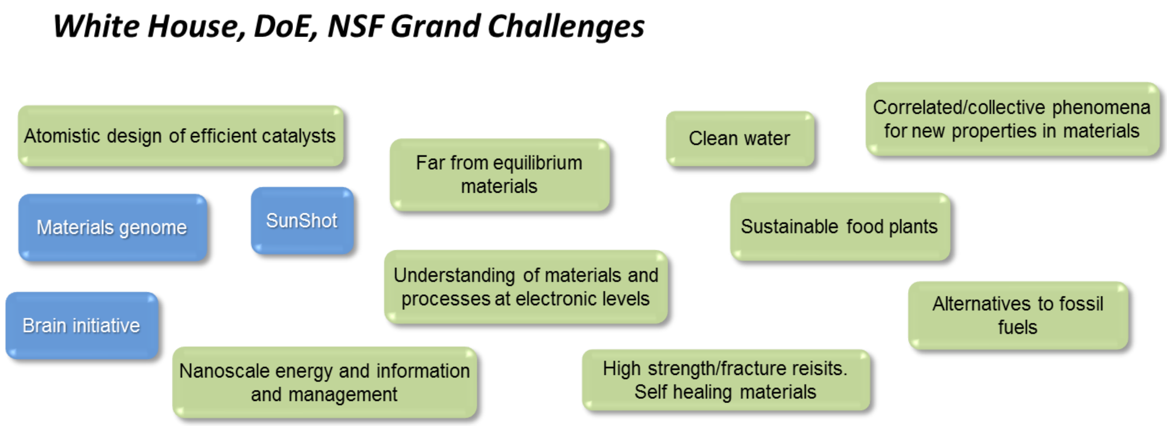


Figure 2-2*. White House, DOE and NSF Grand Challenges in materials and the nanosciences. These are directions for future user activity and therefore need to be part of our vision.*

As a consequence, we have structured our future technical vision into four strategic directions (Figure 2-3): (i) *Energy Efficient Architectures*, (ii) *Nonlinear Nanophotonics and Nanomechanics*, (iii) *Quantum Materials* and (iv) *Computational Nanomaterials*. These four directions fall within our overarching research theme of "*Manipulation of Nansocale Interactions for Energy Efficient Systems"*. These strategic areas span all six of the research groups at the CNM and are discussed in more depth in the accompanying documents that describe the research efforts in the groups. The rationale behind, and overviews of the scientific challenges in each of these strategic areas are outlined below, together with our management plan for supporting them.

The strategic plan was developed following input from our Science Advisory Committee (SAC) and User Executive Committee (UEC), and weekly strategy discussion sessions between the management team that culminated with an off-site "strategy retreat" on February 1-2, 2016 with the participation of the entire division, including technical and management team. The strategic plan emphasized an approach that encompasses our scientific vision as well as provides scientific leadership to bring added value to our users, and in particular attract new user communities. It includes developing expertise and tools aimed at solving scientific challenges in these areas, and thus supports our users at the highest level. Breakthroughs in these directions strongly depend on the integration within crosscutting sciences spanning from electron and hard x-rays visualization to simulation and modeling across multiple time and length scales. We believe that cross-cutting activities, which link the six research groups within the CNM, lead to new science that is greater than the sum of the parts, further enhancing our ability to attract high quality users. Multidisciplinarity and strong interaction across the different groups are therefore a key element of the strategy.

Our strategy is closely linked to Argonne's strategic plan that guide core research programs at Argonne, namely discovery science and engineering, while complementing other DOE user facilities at Argonne, namely the Advanced Photon Source (APS) and the Argonne Leadership Computing Facility (ALCF). We interact with these facilities where appropriate, thereby allowing us to leverage developments across Argonne to enhance the world-leading capabilities available to the CNM users and make available to them a wider range of tools and expertise.

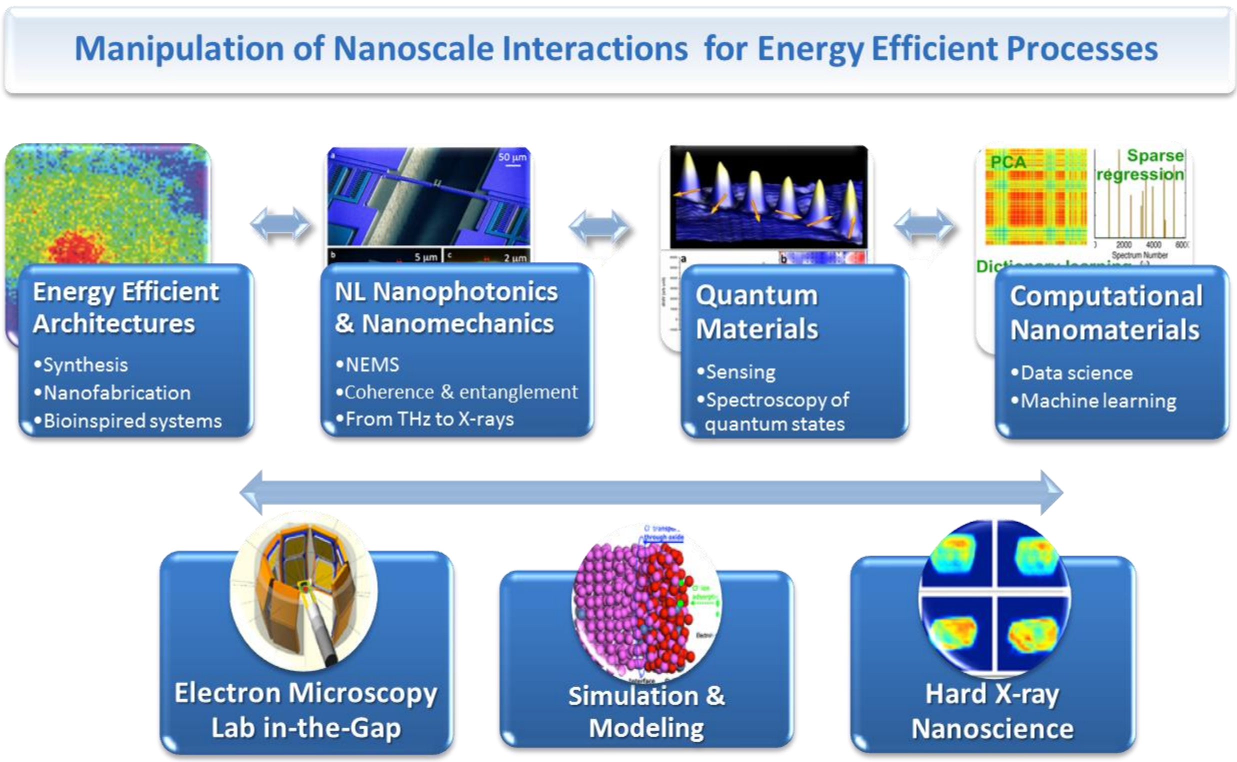


Figure 2-3. *Strategic vision of major scientific direction of the CNM.*

For example, the Hard X-ray Nanoprobe is operated in partnership with the APS with 25% of the beamtime allocated to APS users. Another successful example of new synergistic developments between CNM and APS is a project to build the first dedicated beamline for Synchrotron X-ray Scanning Tunneling Microscopy (SX-STM). In collaboration with APS our Nanofabrication group developed an entirely new class of devices for controlling subnanosecond timing of the delivery of x-rays. This unique capability is being explored for installation at the National Synchrotron Light Source II at Brookhaven National Laboratory. In collaboration with computational scientists at ALCF, the CNM Theory and Modeling group is extending the accuracy frontier in electronic structure calculations. In a recent INCITE grant, we are using a combination of quantum Monte Carlo and multi-reference quantum chemistry methods to improve the description of adsorption energies of molecules on oxide surfaces. The improvement in accuracy will help improve the modeling of photocatalytic and electrocatalytic processes.

The CNM's goal for the next five years is thus to enhance its lead at the forefront of nanoscience and cultivate an advanced user program by identifying scientific directions of critical importance to the user community.

2.1 Description of Strategic Thrusts

The four major thrusts that fall within our over-arching research theme of "*Manipulation of Nansocale Interactions for Energy Efficient Systems"* are described below and are also indicated in the schematic of Figure 2-3. The alignment of the key projects under the strategic thrusts are described (often a project will belong to more than one thrust), as well as the alignment of the project to the NSRC core strengths of synthesis, nanofabrication, characterization and theory, and when appropriate, where a key project is anticipated to develop into a new user capability.

2.1.1 Energy Efficient Architectures (Synthesis, Nanofabrication and Neuromorphic Systems)

A goal of this thrust is to achieve energy efficient systems that utilize hierarchical synthesis and materials design at the nanoscale. It aims at a system level approach that combines synthesis and nanofabrication across different scales and aims to use both self-assembly and top-down approaches (Figures 2-4 and 2-5). The objectives for this approach, in the end, are to identify new pathways in energy transduction including propagation of charge, spin and excitons; new electronic, optical or magnetic functionalities produced by closely coupling nanomaterials with different behaviors; and develop new ways for sensing and achieve adaptive responses to the environment.

The sequential infiltration system (SIS) for instance, is a new film synthesis approach that infiltrates tailored polymers with atomic layer deposited metal oxide precursors to create structures with deliberate porosities that have controllable chemical surfaces and is an example of a system level approach to the fabrication of nanomaterials by combining different techniques. It has high potential for use as energy efficient filtration membranes, and as oleophilic absorbers for oil recovery and cleanup.

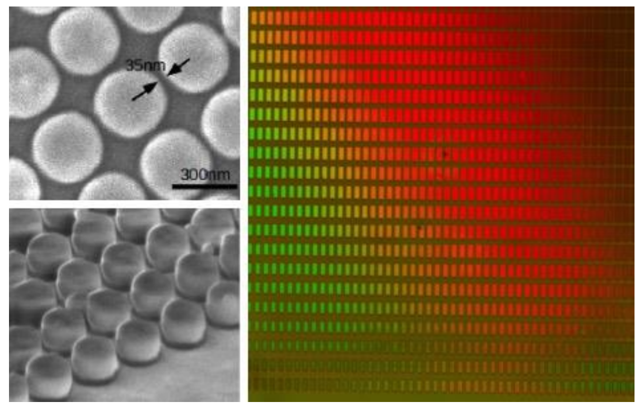


Figure 2-4. *Arrays of dielectric nanoparticles for efficient manipulation of light on nanoscale.*

Within our proposed research program, a few other examples of hybrid nanomaterials under exploration include metal-semiconductor nanostructures to create mixed quantized/collective states for ultrafast nano-optical phenomena, atom-by-atom assemblies of nanostructures made with our scanning probe tools (borophene), and the designed modification of nanoparticle surfaces for catalytic applications.

Our unique characterization approaches will include a new beamline facility, XTIP, which will provide a platform for studying/imaging excited state chemistry at the molecular level, and a rheometer in an x-ray beamline for examining hierarchical flow and shear characteristics in complex nanomaterials at unprecedented resolutions.

With future user capabilities in mind—as a pilot—we will also begin exploration of establishing thin film deposition capabilities directly on the Cloud so that they can be remotely operated by users while providing a “lab-next-door” experience that is secure and efficient. This will be a collaborative project between materials scientists and software programmers. The Cloud has dramatically altered the way we store our data and compute today, and we anticipate that in the next 5-10 years it will dramatically affect the way we do experiments in the future. Putting crystal growth capabilities on the Cloud makes both economic and scientific sense in the way future research could be conducted.

Finally, our strategy in this area will also include nanomaterials and systems for neuromorphic science, where we will look for energy efficient functionality that is inspired by biological transduction and information processing mechanisms. This can have several flavors. This includes the design of materials and controlled defects and inhomogeneities within these materials that can be modified at ultra-low energies (and at a few hundred millivolts) for artificial elements that can be inserted as part of biological circuitry.

This could be elements such as artificial synapses, neurons and axon-like signal propagation elements compatible with biological signal processing and energy delivery. This will also include the integration of various nanotechnologies such as microfluidics, functionalized nanoparticles and chromophores, and nanofabrication to build integrated nanofluidic circuits for crating massively parallel ways for screening,

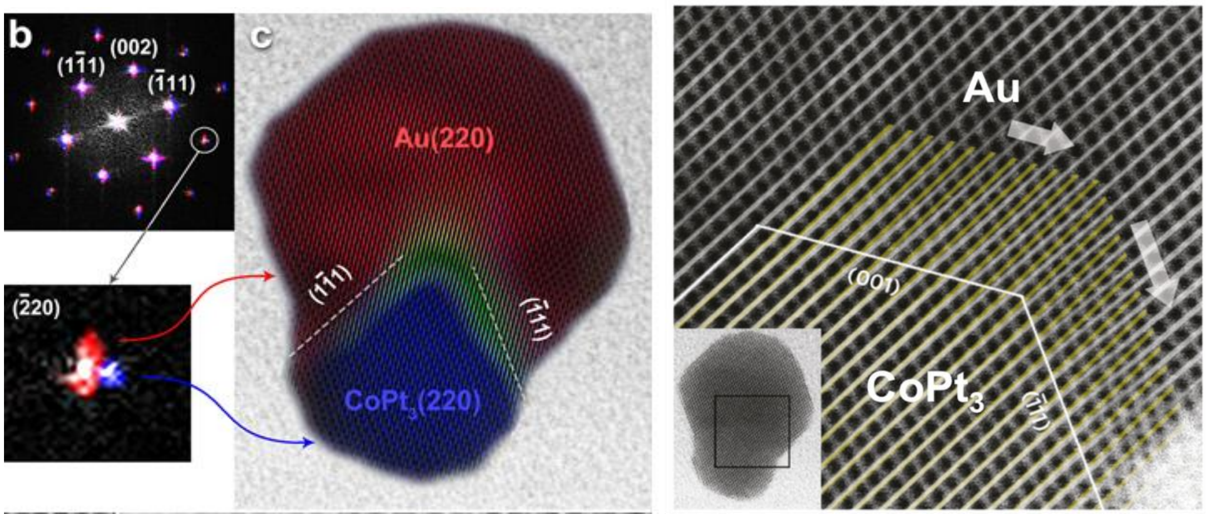


Figure 2-5. *In situ study of the growth mechanism and how the lattice mismatch induces the morphology and controls reaction kinetics of heterostructured NPs*

modifying and analyzing cellular species. On the one hand it enables the use of traditional nanosciences skills to build machines usable for biology (the Drop Seq technique is an example). On the other hand it allows us to use these machines to study energy transfer processes and sensing methodologies that are bio-inspired, which is part of our charter. Finally, on the characterization side, we will look at new ways of using synchrotron x-ray imaging to rapidly image the brain, an area where our traditional skills can bring new value and attract users from the rapidly emerging field of "connectomics".

2.1.2 Nonlinear Nanophotonics and Nanomechanics

Tailoring nanoscale interactions for optical and mechanical response is a frontier in nanoscience that the CNM is unusually well positioned to lead. The impact anticipated from this activity is ambitious and broad and extends from developments in tribology to examining atomistic processes at picosecond scales.

In the CNM, we are developing important new approaches that harness non-linear effects in nanoscale structures. This includes the study of coupled nanoelectromechanical (NEMS) systems whose rich dynamics in the non-linear regime enables new phenomena that has been observed by the team such as frequency stabilization and energy storage via nonlinear coupling (Figure 2-6). It also includes the design of plasmonic structures combined with nanofabrication that allow the creation of flat, microfabricated optical lenses with tailored properties (Figure 2-4). Each of these examples different techniques and approaches, such as photonics, plasmonics, MEMS and self-assembly, to build a new functionality is consistent with the new, more sophisticated direction that the nanosciences need to take. We will continue to study and develop the fundamental science behind manipulation energy—either electromagnetic or mechanical—in such coupled, non-linear systems.

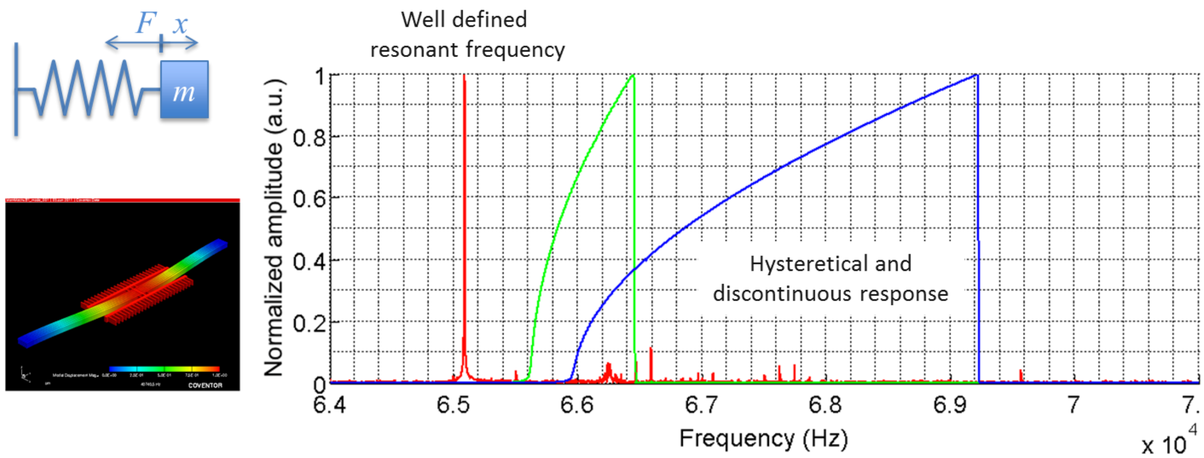


Figure 2-6 *As the dimensions of resonator get reduced toward nanoscale the response became highly non-linear. Non-linearity can be used to enhance the performance of nanomechanical devices to overcome intrinsic limitation. The picture above shows a typical amplitude vs, frequency response of a flexural resonator like the one showed on the left bottom.*

A second area of our strength where we will continue to develop new understanding and new user capabilities is in exploring transient energy transfer, charge transfer and mechanical relaxation phenomena in nanostructures at picosecond scales over a range of materials that extend from semiconductors to biological materials and for applications ranging from catalysis to optoelectronics. User capability enhancements will be in the areas of developing time resolved pump-probe THz capabilities (with scientists from the Advanced Photon Source), which is valuable for a variety of condensed matter phenomena such as phase transitions and coupled spin-and-valley dynamics. There is also increasing interest in combining different fast spectroscopic phenomena together for studyong transient phenomena particularly those related to carrier population evolutions, evolution of structural changes, and their inter-relations in materials subject to stimuli. As a result one of our goals is to put together a capability that combines time resolved photoelectron spectroscopy, cathodoluminescence and electron diffraction again on picosecond timescales, which will offer users new and powerful probing techniques.

2.1.3 Quantum Materials

Quantum materials is a rapidly emerging area rich in opportunities for exploring the physics of entangled states, and for coherent information processing with minimal loss in energy. While most recent interest has been in using quantum materials (such as qubits) for digital (and some analog) quantum computing and communications, the opportunities for using quantum materials for making measurements of materials and surfaces is particularly significant for the NSRCs (Figure 2-7). Quantum sensing refers to: (i) the use of quantum entangled states to make measurements that allow a higher sensitivity than that dictated by classical noise limits, and (ii), using quantum mechanics to extract maximal information from a sensor probing its environment. Development in quantum materials in the past ~5 years warrants the development of user capabilities in this area.

If N classical measurements are made of a physical quantity, the error scales as $1/\sqrt{N}$. On the other hand it has been pointed out that error scales as 1/N for N entangled quantum states. This $\sqrt{N}$ improvement is a driving force behind (i). The demonstration of this is a grand challenge, and developments in qubit design and testing, driven primarily by research in quantum computing, makes quantum sensing a dynamic area for nanoscale measurements for the next five years.

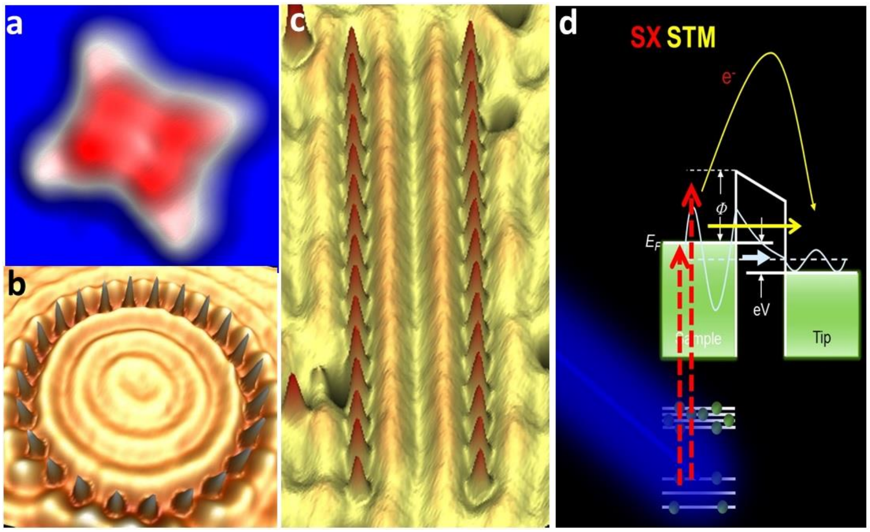


Figure 2-7. a*). STM image of Co-TBrPP molecule.* ***b****) and* ***c****): Atomic quantum corrals.* ***d****) Excitation of core level electrons to the excited states located between the Fermi level and work function of materials in SX-STM, which then tunnel into the tip.*

An example of (i) is in using Noon states, an ensemble of entangled photons, to improve the sensitivity of interferometers compared to classical measurements, where N is the number of photons. An example of (ii) is in using isolated single atomic defects (such as the diamond center in diamond or other defect centers in SiC) to measure minute magnetic fields at high resolution. There is a rich variety of defect states in the large number of insulating oxides and related compounds that are as yet unexplored. Solid state high performance gyrometers, accelerometers and magnetometers that outperform classical counterparts in size, weight and power are other examples of (i).

A grand challenge for quantum materials and sensing is to demonstrate measurements with accuracies below the classical noise limit. This has not been demonstrated. En route to such a demonstration lies many fundamental scientific opportunities that we will pursue—such as demonstrating remote interrogation with entangled qubits, examining how the strength entanglement of two qubits on a surface can be varied via coupling with inserted molecular species, examining the strength of entanglement on the nanoscale as a function of distance (it should not vary) and how one can use the entanglement to detect molecular or atomic analytes. There is also tremendous opportunity in exploiting new materials with controlled and deterministically placed defects and heterogeneities for creating quantum states that would form the basis for such quantum information systems.

The challenge in quantum sensing is to be able to make compact, usable practical sensing systems. Demonstration of robust entanglement, remote readout, and studying the interaction of entangled states with surfaces are areas of fundamental research that can lead to new techniques in quantum sensing and we should plan a strategy to install the first quantum sensing based tools available in a user facility in five years.

2.1.4 Computational materials science--Machine Learning

The exponential growth in computing power and cheap data storage together with the availability of highly scalable atomistic simulation codes have begun to revolutionize the modeling and computational analysis of materials. The ability to simulate larger volumes of a dynamically changing material via different computational techniques is traded off by the time resolution at which the change is monitored. Figure 2-8 shows this trade-off for ab initio, many body, and pair-potential approaches, and charts how this space is expected to increase as we move from peta-FLOP machines to exa-FLOP machines. Concurrently, the advances in computing hardware have also revolutionized data analytics and machine learning, making Big Data a household word that has influenced our lives by its handling of transactional and social data. Going forward, these powerful techniques of data science can play a significant role in computational materials science by combining data analytics with first principles physics. For instance: despite advances in electronic structure methodologies/implementations, there is still a substantial gap between time and length scales

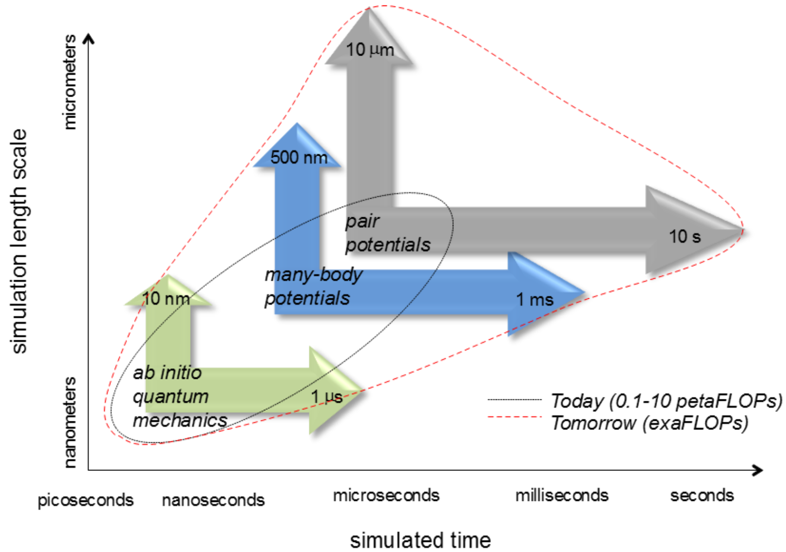


Figure 2-8. *The trade-off space between simulated volumes and time slice resolutions for simulations of dynamically changing materials. Capabilities today (with peta-FLOP computing) are compared against future exa-FLOP computing capabilities. (From A. Curioni, IBM).*

accessible to ab initio molecular dynamics vs. the more computationally efficient but less accurate semi-empirical functional form based classical force fields. To meet this challenge, there is an opportunity to develop a new class of inter-atomic potential functions or force-fields that combine the accuracy and flexibility of electronic structure calculations with the speed of classical potentials by merging and exploiting the best insights from the fields of machine learning, advanced optimization and atomistic simulations. Extensive simulations (such as molecular dynamics simulations for example) throw away a significant fraction of the data because of storage cost considerations and in the end what is stored often may be clouded by the built in bias of the investigator. Streaming data analytics engines can analyze the streaming data from the computer socket prior to its deletion, in order to detect patterns and trends which in the final limit could lead to rule discovery in physics via analytics. Such streaming analysis has been executed with great success in analyzing social and behavioral data and should be extendable to physics. Our vision is to establish a firm foundation for the combination of first principles physics and machine learning/data science over the next five years that will include the establishment of user capabilities that will allow broad use of these techniques. With the improvements in computing power and the introduction of these new techniques a vision for computational materials science should be one that moves towards *prediction* rather than *confirmation* and towards *discovery* rather than *optimization*.

2.1.5 Cross-cutting science: electron microscopy and x-ray analysis

Advancing towards our research goal *Manipulation of Nansocale Interactions for Energy Efficient Systems* requires pushing the envelope in terms of both temporal and spatial resolution for exploring the local behavior of complex nanoscale architectures. Such advances will require increasingly sensitive experiments and sophisticated simulations of nanoscale systems. Advances in visualization capabilities have always resulted in improved understanding of scientific phenomena. Therefore it is critical to expand our horizons by developing crosscutting capabilities that will allow our users' access to new visualization capabilities with improved insight in temporal evolution of events during operation.

The CNM Hard X-ray Nanoprobe (HXN) provides high resolution, multimodal 2D and 3D imaging with diffraction, fluorescence, and transmission contrast modes in a single tool, creating unique opportunities for nanoscience. These powerful capabilities enable visualization of internal structure, chemistry, and morphology – all of which enable in situ studies of materials response to variety of stimuli at a sub-30nm spatial resolution (Figure 2-9). This simultaneous visualization of material properties is particularly well matched to the often complex structure / function relationships in nanoscale materials for energy. Beyond a direct microscopy of materials properties, the HXN also provides more advanced x-ray scattering methods that have the ability to couple to the excitation states of materials – phonons, plasmons, magnetism, chemical bonding, and orbital orientation – enabling a suite of versatile visualization tools for correlated materials. Imaging can be acquired over length scales spanning from nanometers to centimeters which allows the CNM to effectively address emergent science relating to nanoscale and mesoscale hierarchical ordering and fluctuations. Characterization of nanoscale materials using hard x-rays has the fundamental advantage of utilizing a weakly interacting probe that is both nondestructive and penetrating and therefore directly offers a mechanism for investigation of internal states of matter, buried layers and interfaces. As well the structured time resolution of synchrotron x-ray sources for these studies can be harnessed at a nanosecond time regime for time-resolved microscopy and spectroscopy of driven materials out of equilibrium. All of these capabilities are extremely well positioned to exploit the unprecedented brightness and coherence of the APS Upgrade – resulting in two orders of magnitude higher focused flux for the HXN - which will enable entirely new insights into structure-function properties of complex nanoscale systems and carry forward our unique, world-leading contribution to NSRC user studies.

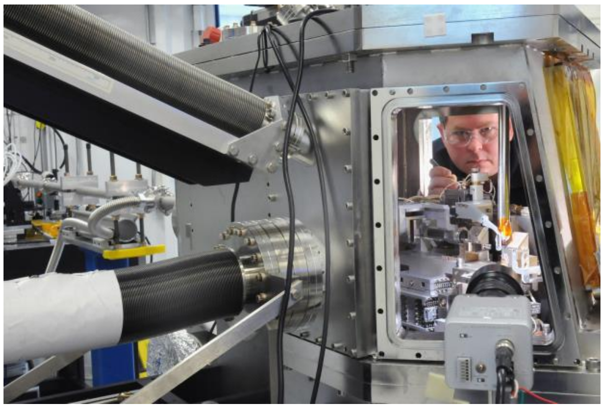


Figure 2-9. *Hard X-ray nanoprobe: 2D and 3D multimodal x-ray imaging with 30 nm spatial resolution extending to 5 nm by ptychography.*

Our recent innovative capability, a MEMS x-ray pulse selector, stemmed out of collaborative research between CNM and APS, is obtained by coupling of x-rays with nanofabrication devices to enable a subnanosecond timing of the delivery of x-rays at synchrotron beamlines. This achievement holds a promise for new understandings of the primary events and how they determine the type of ensuing processes. Collaborative research between CNM and APS also led to another exciting opportunity obtained by coupling of synchrotron x-rays with scanning probe techniques (SX-STM). This new capability extends the reach of CNM hard x-ray microscopy to the atomic scale by enabling atomic resolution imaging of magnetic properties, chemical bonding, and orbital orientation, as well as excited state molecular orbitals relevant for many phenomena ranging from photosynthesis to catalytic reactions and charge and energy transfer processes.

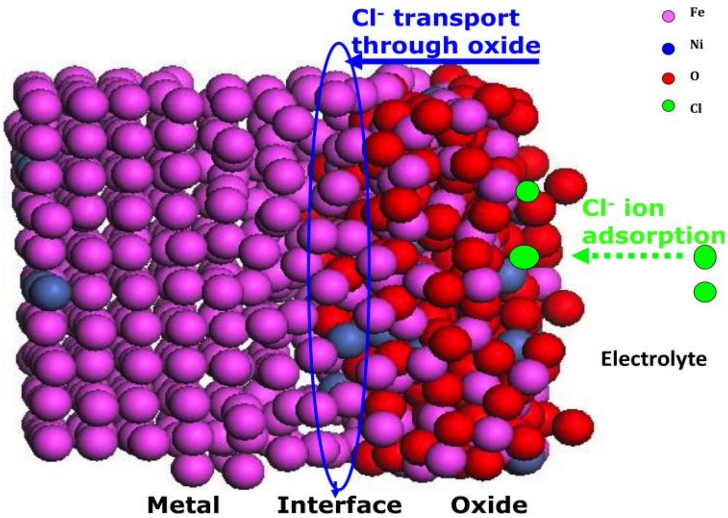


Figure 2-10. *Schematic showing the two key steps believed to lead to pitting corrosion (formation of voids): chloride ion adsorption on to the oxide surface and chloride ion transport through the passive oxide film.*

Our theory and modeling capabilities provide cross-cutting support for many efforts within the CNM. The state-of-the-art electronic structure, molecular dynamics, and electrodynamics simulations software, developed in the CNM and implemented on its high-performance computing cluster, enable the interpretation and prediction of a wide range of experimental results generated by the CNM's staff and user community (Figure 2-10). The group's growing data science/machine learning component will further enable the scientific discovery process.

Electron microscopy capabilities provide another complementary cross cutting capability that enables high quality of our science. A unique component of our research in transmission microscopy capabilities is the application of chromatic aberration correction in electron microscopy. We host one of only a few instruments world-wide with chromatic aberration correction. This recent development in electron microscopy allows us to exploit high-resolution amplitude contrast imaging to provide atomic scale elemental and structural information in one directly interpretable image (Figure 2-11). It also provides ability to perform high-resolution structural characterization in complex systems, capable of distinguishing even Ca and Ba in their complex oxides for example. This unique capability will enable a variety of research activities that address issues related to interfacial and surface structures for which electron microscopy is one of the only techniques that can provide such information.

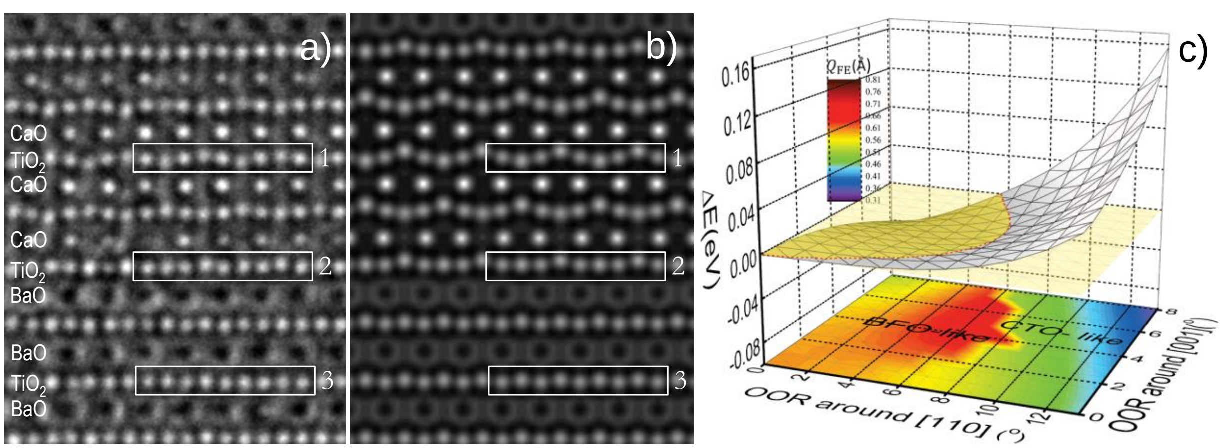


Figure 2-11 *Experimental a) and calculated b) HRTEM images showing oxygen octahedral tilts in a 4BTO/4CTO superlattice film grown on an STO substrate along [110]. Simulated HRTEM image using the atomic positions obtained from first principles calculations. c) Metastable CaTiO3 polar phase with a characteristic of BiFeO3 is stabilized (in red area).*

A new exciting class of experiments using electron microscopy encompasses visualization of the dynamic response of a material to an externally applied stimulus. In situ microscopy is well suited for direct visualization of nanoscale interactions in their natural or operating environments. These new capabilities can visualize structural changes during battery cycling, or changes of catalysts atoms upon adsorption of reactive molecules. At the same time, building on our core strengths in spectroscopy, we are working to enhance the capability for spectroscopy during in situ experiments. Our long-term and largest development activity is aimed toward "Lab-in-the-gap", starting with the development of a large-gap instrument with exceptional dynamic capability. This capability will include the design of an Inverse Hyper-Spectral Imaging (IHSI) system that will combine the spatial resolution of electron probes with the high energy resolution of optical spectroscopy enabling structure-function determination of complex materials. This activity will be a collaboration among the electron microscopy programs at all five of the NSRC's and will build on expertise at each of them and from across the scientific community.

Appetite for risk: many of the projects in our strategic plan are ambitious and our strategy here possesses a "high-risk-high-payoff" element. We have aimed for a high bar here—there is a great deal of uncertainty and unknowns in where we are headed. Projects such as brain imaging using x-rays, and building entangled qubits for measuring surfaces are difficult and challenging projects with many unknowns. However our vision is one where we assume a leadership role to not only provide the user capabilities that our users will need 2-5 years from now, but also to lead and identify the areas of the nanosciences where research needs to be done.

2.1.6 Key staffing decisions

As we embrace the emerging new directions laid out in the vision section, we intend to make strategic hires across the CNM that will support our strategy and enhance the capabilities, expertise and support that we can make available to our users. The planned new hires include both junior and midlevel scientific PI staff, with all of the planned hires providing a very high level of support to the user program

3. Development of Synergies with other DOE User Facilities at ANL

The CNM powerfully leverages it science and its capabilities with the strengths of the other BES user facilities co-located at Argonne National Laboratory. Examples of such synergies are provided below.

3.1 Hard X-ray Nanoprobe Facility

The CNM's Hard X-ray Nanoprobe facility was jointly built and is operated by CNM and APS. It is the only dedicated x-ray microscopy facility within the portfolios of the nation's five Nanoscale Science Research Centers and its nanoscale imaging capabilities are without peer worldwide. The CNM X-ray Microscopy Group directs the scientific program, manages the operations, and provides the majority of the funding for the Hard X-ray Nanoprobe. APS provides scientific and technical support to the Hard X-ray Nanoprobe through an in-kind effort contribution. Experimental beamtime at the Hard X-ray Nanoprobe is allocated on a 75% - 25% basis between CNM and APS. Proposals for beamtime may be submitted either through the APS or CNM proposal submission portals.

3.2 Dedicated Beamline for Synchrotron X-ray Scanning Tunneling Microscopy

A successful example of the CNM synergy with APS is the recent development of the world's first dedicated SX-STM beamline, called "XTIP", at the Advanced Photon Source. Chemical x-ray imaging with atomic and nanoscale resolution has been a long-standing goal since the STM was invented. The CNM and APS scientists recently teamed up to address this challenge and develop a new technique that aims to simultaneously resolve chemical element speciation and topology of nanoscale materials down to single atoms and molecules (Figure 3-1). The SX-STM combines two of the most powerful techniques for materials characterization, namely synchrotron x-rays (SX) and scanning tunneling microscopy (STM), into a single instrument that is intended to enable exceptional resolution for nanoscale imaging with elemental, chemical, and magnetic sensitivity. Our critical innovations of a "topological filter" and nanofabricated "smart tips" enabled us to overcome experimental barriers and demonstrate (in 2014) for the first time, x-ray chemical imaging at atomic limits. The current project was in part inspired by these developments. Recognizing the technique's potential, the US DOE Basic Energy Sciences Advisory Committee (BESAC) highlighted this technique as an important approach in future research roadmap in their recent report on Transformative Opportunities for Discovery Science. The new beamline is being built at APS Sector 4 to take full advantage of the brightness and polarization control of the undulator source there. XTIP will be completed in 2019; the SX-STM program will receive 20% of the beamtime at beamline 4-ID in the interim, during the XTIP construction period.

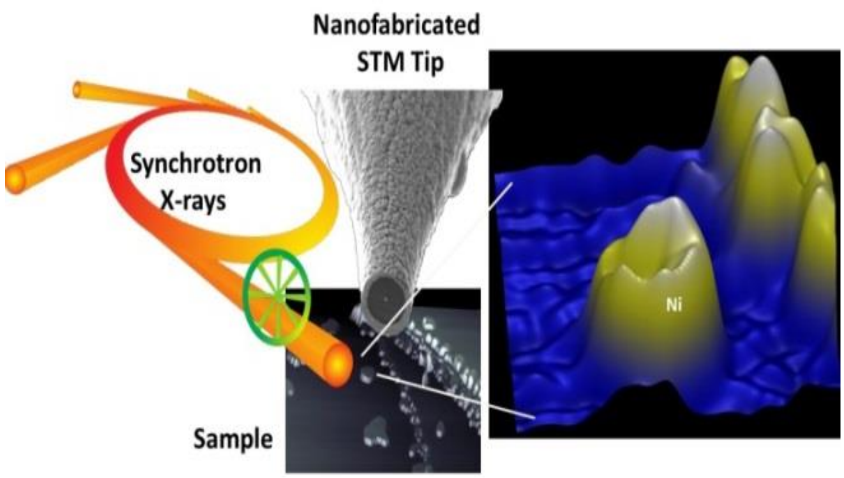


Figure 3-1. *An exemplary success story from a close collaboration within Argonne is the development of SX-STM that combines two powerful characterization tools, enabling the imaging of nano-structures and their chemical and elemental compositions simultaneously at atomic limits.*

3.3 MEMS X-ray Pulse Selector

Recent collaboration between CNM and APS also led to the work that resulted in a *MEMS x-ray pulse selector*, a new class of devices for controlling subnanosecond timing of the delivery of x-rays. Shrinking of x-ray optics to the microscale using MEMS technology, created an opportunity for developing ultrafast devices that reflect x-rays at precise times and specific angles. This work may lead to compact, sophisticated x-ray optical approaches for studying the structure and dynamics of matter at atomic length and ultrafast time scales (Figure 3-2). Recent experiments demonstrate that these devices can achieve sub-ns gating windows (~500 ps). This goes far beyond what any state-of-the-art competing technology, i.e. mechanical spinning disc choppers, can do by ~2 orders of magnitude. Although MEMS technology has found a wide range of photonics applications in industry and basic research, MEMS developed for dynamic x-ray manipulation is a new idea. This research demonstrates the potential for a compact, scalable, low-cost and reliable solution to control the beam and pulse train from current and future advanced x-ray sources. In analogy to MEMS photonics devices for the visible and telecom wavelengths, this work is expected to lay the groundwork for the development of a suite of x-ray optics, e.g. ultrafast gating devices, multiplexers, ultrafast spectrometers/monochromaters, to facilitate experiments currently not possible at x-ray synchrotrons.

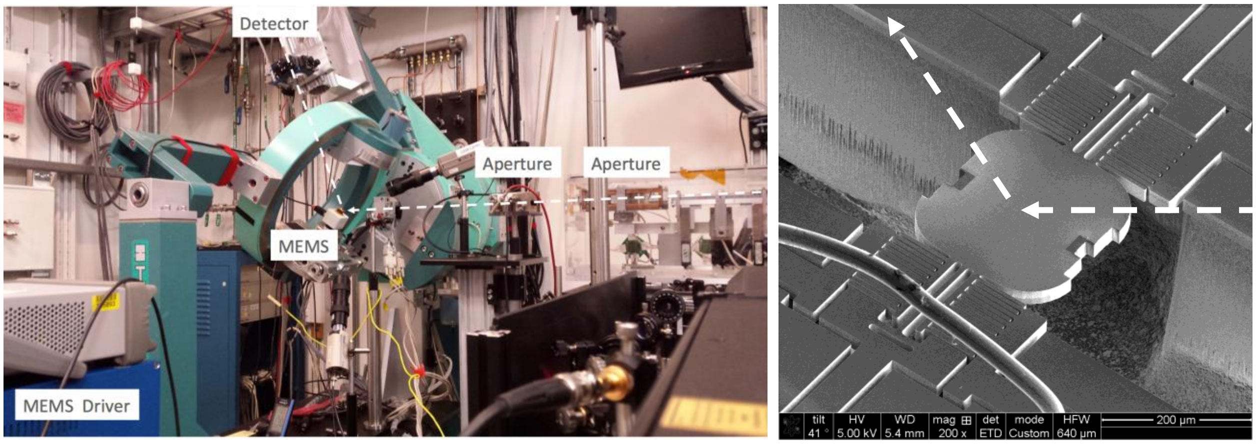


Figure 3-2 *Ultra-fast gating of x-ray pulses from synchrotrons (left) using MEMS devices (right)*

3.4 Integrated Computational Tools

The *Theory and Modeling Group* has strong associations with Argonne's Leadership Computing Facility (ALCF). For example, DOE INCITE projects entitled "Mesoscale reactive simulations of electrochemical interfaces" and "Combining high accuracy electronic structure methods to study surface reactions" are part of CNM's research portfolio. These projects involve significant amounts of computer time on some of the fastest supercomputers in the world and also involve working closely with ALCF staff members on developing and improving the performance of molecular dynamics codes (LAMMPS and NAMD) and on quantum Monte Carlo electronic structure approaches. Members of the ALCF also participate in the *Theory and Modeling Group's* group meetings.

3.5 Integration of the Electron Microscopy Center

A key activity during FY 2015 that enhanced available capabilities for our users was the integration of the EMC within the CNM. This integration expanded our scientific synergies, of which there were already many, and made users' access to a broader set of tools and expertise easier via a single user-proposal system. We started the integration by transferring the EMC into the home division of the CNM, the Nanoscience and Technology Division (NST), during FY 2014. With this transfer, major functions of the EMC including management, safety, IT, and building operations were administered within NST. The full merger of the EMC's proposal system was established in FY 2015. Beginning in March 2014, EMC capabilities were included on the CNM user proposal form; this meant a user could access both facilities on the same project for a short time prior to full EMC integration. Analysis of the number of users for FY2015 showed an increase of 17% unique users compared to previous years, primarily as a direct result of the integration of the EMC.

3.6 Partnership with the Laboratory

Argonne National Laboratory is in the process of building an additional *6000 sq ft of cleanroom space* that will be connected to the CNM's existing cleanroom (Figure 3-3). The extension of the existing cleanroom will house prototyping and testing equipment to enable the advancement of nanoscale devices from the research phase to development. This option is expected to be attractive to industrial users interested in developing nanofabrication processes compatible with commercial semiconductor technology. Examples of possible instrumentation include: steppers for large area lithography, flip-chip bonders for heterogeneous integration, and packaging tools. Construction is planned for May 2016 through May 2017.

Argonne is also in the process of constructing a *liquid helium recovery system* in order to recover helium gas currently being lost from the boil off and helium transfers of cryogenic instruments. The CNM will have a recovery & compression facility that will transport captured helium to the liquefaction plant located in the

Physics division, the largest consumer of helium on site. After liquefaction helium will be delivered back to CNM for recycled use. Installation of the helium recovery facility is expected in 2017.

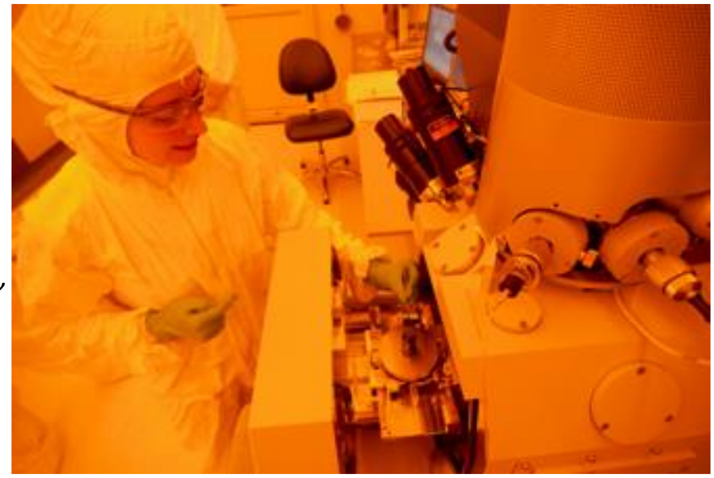


Figure 3-3. *CNM nanofabrication facilities include a fully equipped cleanroom [class 100] with electron beam and optical lithography, FIB, a wafer stepper, deposition, metrology, and etching capabilities.*

4. Crosscutting Research with other Programs

4.1 Crosscutting Research with Core Research Programs at Argonne

Science and capabilities at the CNM leverage core research strengths with several research programs at Argonne. For example, the CNM Theory & Modeling group is a participant in the Center for Electrochemical Energy Science (CEES, a DOE Energy Frontier Research Center), the Midwest Integrated Center for Computational Materials (MICCOM, a DOE-Basic Energy Science funded Center), SunShot Bridging Research Interactions Through Collaborative Development Grants in Energy (BRIDGE, a DOE-Energy Efficiency and Renewable Energy program), and a Strategic Partnership Project (SPP) with Toyota Research Institute of North America (TRINA). In CEES, we have developed computational approaches to configurational sampling for non-equilibrium electrochemical processes in high capacity lithium-ion and beyond-lithium-ion energy storage materials, and a computational capability for modeling Raman spectra. In MICCOM, we are developing scale-bridging capabilities for the modeling of solid-liquid interfaces and thermal transport in nanostructured materials. In the BRIDGE project, we are developing a high throughput computational framework for sampling and evaluating grain boundaries in semiconductors. As part of the SPP with TRINA, we have developed methodology to study the electronic and thermal transport properties of metal-insulator materials. These new capabilities strengthen and expand the intellectual and scientific expertise that is available at the CNM.

Our expertise in time resolved spectroscopy has contributed to a DOE BES ultrafast science project that aims to determine impacts of coherent phenomena on electronic processes such as molecular motion-driven photocatalyst activation. In this project the CNM is making time-resolved optical measurements of nanomaterials that exhibit coherent vibrational motion and correlating them with electronic phenomena such as energy transfer. The results aim to connect to nonequilibrium energy flow and to photocatalysis,

The CNM is collaborating with a DOE BES Materials Sciences Program with the Institute for Molecular Engineering (IME, a partnership between the University of Chicago and Argonne) entitled "Quantum Metamaterials". The intent of the collaboration is to establish a vibrant quantum materials and sensing research effort within Argonne, and to benefit from the deep expertise that currently exists at IME on the subject. The goal is to explore the physics of quantum states and entangled states for energy efficient information processing. New developments in qubit and quantum material development resulting from research on quantum computing research have made this a timely research topic, though in the case of our work, **the focus will be on utilizing such materials and devices for sensing and measurement instead of digital computing.** Candidate systems include solid-state color centers and deliberately created defects in insulators (such as the NV center in diamond), correlated optical photons, GHz acoustic phonons, and microwave photons in superconducting structures, all areas in which our team has significant expertise.

Sequential infiltration synthesis (SIS), a novel materials synthesis technique invented and developed at CNM in partnership with researchers in the Energy Systems Division at Argonne, has generated interest in diverse research communities. Within Argonne, SIS has been leveraged in programs supported by the U.S. Coast Guard for oil spill remediation, JCESR for novel materials for energy storage, and the IME for functionalization of polymeric materials. Moreover, a budding program at Argonne built on materials science and engineering for water research will fold SIS techniques into many topical studies for fluid separations and membrane science. Beyond Argonne, SIS has been adopted in active research at IMEC for next-generation lithography technology, at other NSRCs such as CFN, and in other industrial research laboratories.

4.2 Partnership with other NSRCs

The EMC group is working together with the electron microscopy groups of the other NSRC's to identify and develop the next generation instruments that create new scientific opportunities and advance the field beyond off-the-shelf technology. Building on guidance from the BES workshop report "Future of Electron Scattering and Diffraction", we plan to exploit aberration correction to develop a "lab-in-the-gap" instrument that will transform the electron microscope from what is primarily an imaging instrument into a flexible nano-laboratory (Figure 4-1). This activity will build on expertise at each of the NSRC's and will also include experts from across the scientific community.

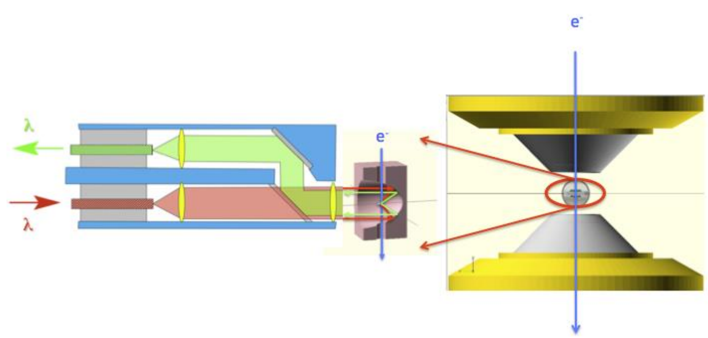


Figure 4-1. *Proposed LG-S/TEM electron/optical hyperspectral system. Using parabolic and associated transmission/reflection mirrors, lenses and light pipes, the specimen can be illuminated while simultaneous photo and cathodoluminesce signals are generated and/or detected.*

4.3 Partnership with Users

Several partner user proposals (PUP) are in place at this time with various Argonne divisions. For example, the CNM is developing time-resolved terahertz (THz) spectroscopy that leverages a femtosecond amplifier in the CNM laser labs, while the APS funds the construction of a THz spectrometer. THz spectroscopy is valuable for studying a large range of condensed matter phenomena, including, for example, phase transitions in complex oxide nanomaterials and coupled spin and valley dynamics in two dimensional transition metal dichalcogenides. When complete, CNM staff and users will gain a new, cutting-edge capability for characterizing nanomaterials at THz frequencies including THz time-domain spectroscopy, THz-pump THz-probe, THz-pump optical-probe, and optical-pump THz-probe. These capabilities will complement newly developed THz pump x-ray probe capability at the APS.

A PUP with Argonne's High Energy Physics division involves a research and development effort with the CNM to fabricate large arrays of multi-chroic Transition Edge Sensor (TES) bolometers for use in Cosmic Microwave Background (CMB) experiments (Figure 3-5). The focus is on developing techniques to implement and control the lateral proximity effect and to reduce two-level-system loss in superconducting microstrip at mm-wave frequencies. The goal is the stable and robust production of multi-chroic microstrip-coupled TES bolometer arrays across multiple 150 mm substrates. Several tools installed in the CNM clean room as part of this PUP are available for users (an ASML stepper, an etcher, and two AJA deposition tools).

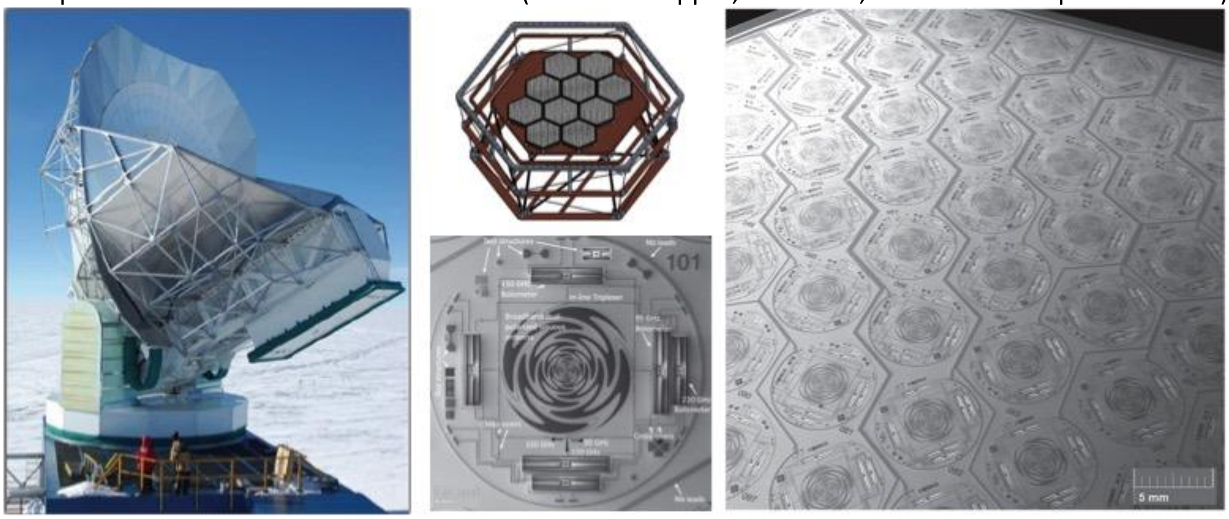


Figure 3-5. *Clockwise from left: South Pole Telescope where the next-generation CMB polarization experiments are performed; CAD drawing of the SPT-3G focal plane and support structure; SEM micrograph of a fabricated SPT-3G multichroic pixel array, including the wiring layout; and SPT-3G multichroic individual pixel.*

The Materials Science Division has a PUP with CNM to complement their research program on new classes of complex oxide thin films, superlattices and heterostructures with single atomic layer control, where novel states of condensed matter may be realized and manipulated with external fields. The materials are synthesized using oxide MBE based techniques. The goals of this program are: (i) Tailoring dipolar interactions in oxide materials to control electronic and magnetic properties (ii) Tailoring 'redox' charge transfer at oxide interfaces, particularly between metallic oxides, to create novel interfacial states, and (iii) Creating thin films with charge-ordered and vacancy-ordered structures for studies of resistive switching using THz and ionic-liquid gating.

A second PUP with APS is to utilize the HPC Carbon cluster for finite element simulation. The APS-Upgrade project will replace the APS storage ring with a new multi-bend acromat (MBA) lattice to improve beam quality; providing ~1000 order-of-magnitude improvement in brightness and coherent flux. The PUP allows APS access to the CNM licensed COMSOL finite element software for development of capabilities that can help APS to design better scientific instruments to enable nanoscale research. APS enhances COMSOL capabilities by adding modules that allow APS and the CNM user community to use multiphysics simulation capabilities. Some projects include nanopositioning for x-ray optics and samples, design of an acoustic levitator to dispense nanoliter samples, ultra high heat flux front-end and beamline components, design of mirror bending and focusing using piezo-electric actuators with nanoscale actuation, and design of a nano size beam analyzer.

A partner user proposal with APS encompasses hard x-ray polarizers and dichroic coherent diffractive imaging for development of a new capability for imaging nanomagnetic stucture simultaneously with crystal lattice strains. This new capability will enable, for example, probing the relationship of magnetic to lattice ordering in engineered nanocrystals and nanostructures films, orbital and charge ordering in the doped manganites, and lattice-spin coupling in multiferroic systems. This project advances our goals to develop dichroic coherent imaging capabilities at the CNM Hard X-ray Nanoprobe and SX-STM programs at beamline 26-ID, and to position the Nanoprobe to take full advantage of the unprecedented 100-fold increase in coherent flux to be provided by the APS Upgrade. In future this methodology will be extensible to study of dynamics of magnetic and other systems by x-ray pump-probe methods and with ultrafast x-ray laser sources.

4.4 Industrial Outreach

One of the goals of the CNM is to increase industrial user participation in order to more fully embrace nanotechnology aspects and relevance to applied technologies. Key long-term industrial partners have included researchers from large companies such as Corning, Inc., Toyota Research Institute of North America, IBM, HP, GE Global Research Center, and small businesses such as Creatv Microtech, Advanced Diamond Technology, and OptoNet, Inc. Awareness of the CNM's user capabilities within the industrial community has expanded recently to include Toyota Motor Engineering & Manufacturing North America, Inc., Brewer Science Inc., several small businesses, as well as the integration of industrial users of the EMC such as BP and UOP LLC; the latter are large oil companies primarily interested in catalyst development when accessing EMC and CNM. The CNM's industrial users have accessed all aspects of our capabilities from the HXN beamline to the nanofabrication facilities, and from various microscopy techniques to the high-performance computing cluster. The CNM maintains an "Information for Industrial Users" page on the CNM public website which includes industrial research highlights as examples for industrial scientists to benchmark their needs to what CNM can offer (http://www.anl.gov/cnm/user-information/industrial-users).

5. Key Instrumentation Decisions

Our objective in prioritizing our recapitalization and facility upgrade plan is to: (i), be able to intercept user needs for the future and to engage users in a program that reaches beyond traditional CNM capabilities; and (ii), allow us to successfully pursue the four strategic areas that we have identified (described in Section 2) and which will provide scientific leadership, attract new user communities and bring added value to our users.

The CNM has a formal review process for prioritizing equipment purchases for equipment value >$50K, while lower-value equipment is left to the discretion of group leaders group leaders, the Associate Director for User Programs, the Deputy Director, and the Director. An equipment plan is evaluated against the recommendations of the UEC, impact to the user community, recommendations of the SAC and strategic vision. The prioritized capital list remains a living document and is regularly updated.

Input for creating the CNM equipment plan has, and will continue, to come from several sources, as described below:

- Input from user community and CNM staff, through CNM-sponsored focused workshops, the annual CNM Users' meeting, strategic science retreats and town hall meetings for the CNM staff, and via informal interactions with staff and users.
- Suggestions from the user community via the CNM User Executive Committee
- Identification of user projects requiring capabilities that do not yet exist in the CNM; also of user demands that exceed available resources.

- The CNM seeks advice from the SAC on a regular basis. The SAC continues to provide valuable input on future scientific directions and on resources that are needed to support these scientific directions.

There is a close tie between our strategic plans for equipment acquisition and CNM staffing plans for FY16–FY18, both of which tie in with our scientific visions for the future, presented in section 2. These strategic areas have been chosen because we believe they will be of critical importance to our user community and will address the key challenges of nanoscience and nanotechnology in the future. Our prioritized plan includes both replacement of existing capabilities to keep them cutting edge, and addition of new capabilities that will enhance the facility's attractiveness to new users, as well as provide differentiation from the other NSRCs.

We will work closely with the Advanced Photon Source to ensure that our strategic directions for development of the HXN at the Sector 26 beamline are synergistic with the MBA lattice envisioned for the APS Upgrade, and the two-orders-of-magnitude greater brightness it will deliver. This will directly translate to 100x greater nano-focused flux on a sample, enabling transformative science with the scanning probe HXN instruments. Nano-resolved studies of spin, charge, and orbital ordering in systems such as complex oxides will become feasible in scanning nano-diffraction mode, as will trace element analysis in nano-biological systems in scanning fluorescence mode.

Instrumentation developments that we plan to pursue at the CNM include:

- Argonne is leading the world in development of SX-STM that combines two of the most powerful techniques for materials characterization, synchrotron x-rays and scanning tunneling microscopy, into a single instrument. Integration of two approaches enables exceptional resolution for nanoscale imaging with elemental, chemical, and magnetic sensitivity. We are currently building the world first beamline dedicated for SX-STM, "XTIP", at Sector 4 of the Advanced Photon Source. This new undulator beamline offers circularly polarized x-rays and an energy resolving power of 4000 over the 450-1600 eV soft x-ray range.

- Upgrades to our unique HXN facility including capabilities for multimodal 3D imaging via fluorescence and tomography, in-situ temperature and electric/magnetic field manipulation, implementation of a diamond phase retarder for generation of polarized x-rays for nanomagnetism experiments, and new zone plate lenses with 20 nm resolution. Together, these improvements will enable us to take full advantage of the unprecedented 100-fold increase in the source brightness of the APS Upgrade.

- Recapitalization of the tools in the cleanroom is of essence for CNM and our user program. Deposition of the thin films of different materials is the very important step in device fabrication flow. E-beam evaporation technology enables depositing ultra-pure films of materials that are difficult to deposit by thermal evaporation, such as dielectrics and high-melting-point metals, with high throughput, sequential coating capability and excellent deposition flexibility. E-beam tool is an ideal solution for deposition of thin films for lift-off process and offers precise thin film deposition rate control, excellent material utilization, freedom from contaminations, and precise film composition.

- The advance spectroscopy during in-situ characterization will open several new research avenues at the EMC and CNM. New instrumentation that enables expanding spectroscopic and tomographic capabilities is expected to enable quantitative materials characterization in multiple dimensions with high-resolution of S/TEM and TEM imaging with EDS performance, including unique EDS tomography. This new capability will benefit practically all CNM and EMC users.

- We are developing a capability to perform single photon detection in the near-infrared spectral region. This capability will actually use two superconducting nanowire single photon detectors (SNSPDs) to enable determination of the correlation function between single photons emitter from two coupled sources. This is a necessary tool for pursuing quantum entanglement studies, and couples strongly to the new quantum materials and sensing strategic direction of the CNM.

- Development of novel scanning probe tools and techniques that allow user research at milliKelvin temperatures and under high magnetic fields, tailored to the energy resolution necessary for exploring quantum phenomena. Proposed research in quantum materials and quantum sensing involves manipulation of spin behaviors at ultralow temperatures. We are proposing development of a mK temperature (less than 50 mK) scanning tunneling microscope coupled with an internal magnetic field strength of 9T or higher within the next 5 years. This system will also be the lowest temperature STM available for the users among the NSRCs.

- The CNM computing cluster facilitates a significant fraction of the CNM user science. The cluster is a combination of nodes from three acquisition phases, the earliest of which were procured six years ago. Over the course of the next two to three years we plan to replace Infiniband switch and older computer nodes and storage with current hardware, thereby increasing our compute capacity while lowering power consumption.

- Optical parametric amplifier (OPA) with a narrowed spectral width and a picosecond (ps) pulse was recently installed. The ps-OPA offers tunability from 470-2400nm and can be implemented in a variety of experiments. These range from transient fluorescence line narrowing to time-resolved stimulated Raman measurements. These measurements are important for establishing, for example, the specific phonon modes and their dynamics that transport heat in nanostructures, as well as for exciting and probing subpopulations of nanostructures in an inhomogeneous distribution of nanoparticles.

- Development of time-resolved combined capabilities of transient photoelectron spectroscopy (trPES), time-resolved electron diffraction (trED), and time-resolved cathodoluminescence (trCL) will enable direct monitoring of the temporal and structural evolution of excitonic interactions and charge separation in hybrid nanomaterials. While spectroscopic techniques offer high temporal resolution, the effect of the structure response to the excitation cannot be elucidated by spectroscopic techniques alone. The proposed combined approach Spectroscopic, Ultrafast Capable, Combined Electron Excitation/Emission/Diffraction (SUCCEEED) would enable for the first time to incorporate the effects of structure evolution and how it alters the landscape of excited states into complete description of the entangled non-equilibrium electronic and structural states.

5.1 Areas of Particular Need of Long Term Recapitalization

Nanofabrication: We believe that it is critical to keep our nanofabrication capabilities state-of-the-art. The nanofabrication facilities in the cleanroom are heavily utilized by users (over 350 usages per year). In order to continue providing our users with timely and efficient access to the best nanofabrication equipment, we need to upgrade existing lithography capabilities.

- The JEOL 9300 electron beam lithography system is the most popular tool in our cleanroom. This system is nearing ten years of continuous use and will require a significant upgrade in order to provide reliable and reproducible sub-10 nm patterning capabilities. An upgrade to a state-of-the-art JEOL 9500 system (2 nm beam size or better) would be ideal. Also, tools ten years or older often cannot be covered under a service contract, hence their maintenance costs become prohibitive high.

- We have recently incorporated a used ASML step-and-repeat lithography system that will help to accommodate the needs of users requiring wafer scale patterning of micro and nanostructures in the short term. While the current system will support a large number of projects with critical dimensions around 500 nm, a more precise stepper, possibly a DUV 248 nm laser based, capable of large scale patterning of features close to 200 nm will be essential to achieve very large scale integration (VLSI) of nanostructures on a 5 year timescale. Furthermore, since step-and-repeat lithography is the standard patterning technique used in the semiconductor industry, availability of this technology will be attractive to users interested in commercializing nanotechnology.

New multiplexed capabilities in spectroscopy: Next generation electron-photon spectroscopies for high spatial resolution and sensitivity can be achieved in a large-gap lens configuration that can accommodate enhanced multimodal capabilities and upon which fast sources and detectors can be incorporated. Many materials issues require this level of characterization that goes beyond current capabilities. For example, the structure and local chemistry of defects and boundary structures have a strong effect on the efficiency of photovoltaics, including the recently discovered high efficiency perovskite solar cells. Composed of mixed organic and inorganic components that can be highly unstable under an electron beam, these materials pose exceptional challenges for temporal and chemical sensitivity for localized characterization that new, high efficiency spectroscopies can address.

The microscopy programs in the five the NSRC’s propose to work together to provide this capability by developing a “lab-in-the-gap” instrument with outstanding dynamic capabilities. This project will build on expertise at each of electron microscopy programs in the NSRC’s and from across the scientific community. As part of the CNM component of that effort, we will design and build an Inverse Hyper-Spectral Imaging (IHSI) system that will combine the spatial resolution of electron probes with the high energy resolution of optical spectroscopy.

This instrument will combine innovative detector technology and new large gap lens design to achieve picoscale imaging and world class spatial resolution and sensitivity in x-ray and electron spectroscopy. All of these are of enormous importance to characterization of nanoscale materials. This instrument will also provide powerful capabilities for high-resolution 3-D electron tomography not only for structure, but also for chemistry. This capability is highly complementary to CNM’s hard x-ray nanoprobe for volume scattering and fluorescence studies of individual isolated nanoparticles and will provide new opportunities for correlative microscopy (electrons and photons) that will allow us to bridge length scales from the atomic scale to the mesoscale.

The development of this capability will be carried out in collaboration with major instrument manufacturers, and the culmination of this project will be a new instrument with world-class capabilities for electron-photon spectroscopies with high spatial resolution and sensitivity. Our user community has consistently identified this configuration as one of its highest priorities and it provides the capabilities that are most often requested by industrial users. In addition, the capability for remote access creates the opportunity to attract new users from the broader scientific community.

6. User Program and Outreach Activities

The CNM user program continually strives to attract the highest-impact users possible, including researchers from across the country and around the globe. Figure 6-1 displays the diversity of CNM users as a function of their affiliation, showing nearly half of the 500+ users per year are from U.S. academia, with lesser percentages from industrial, other government, and international organizations. Figure 6-2 provides the specific affiliation for the non-Argonne users as a function of the CNM group with which they interact; the affiliations are grouped as industrial, academic, government, and international.

The number of refereed journal articles is one of the primary quantitative metrics of project success at the CNM. The number of such journal articles is shown in Figure 6-3 and includes articles by both CNM staff and users. DOE has provided the NSRCs with a list of the top 20 highest impact journals for publishing nanoscience or nanotechnology results. Figure 6-3 summarizes the results by year and by the percentage of high impact journal articles published during 2013-2015. On average during this three-year timespan, nearly one-third (31%) of all CNM journal articles appear on the NSRC high-impact journal list. There was a significant increase in the total number of articles published in 2015, from approximately 230/year to 280/year, most likely as a result of the integration of the EMC.

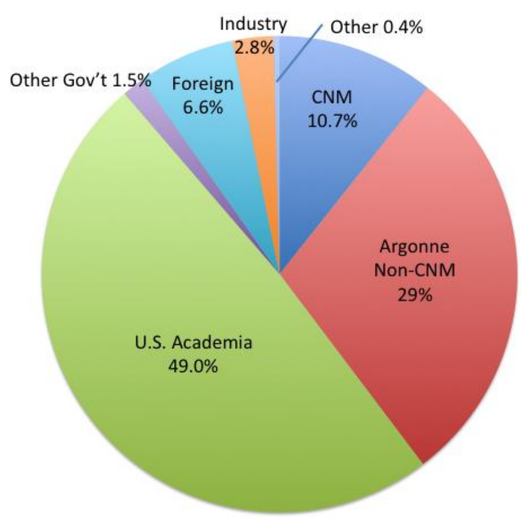


Figure 6-1. *Institutional affiliations of CNM users by affiliation during FY2013-2015.*

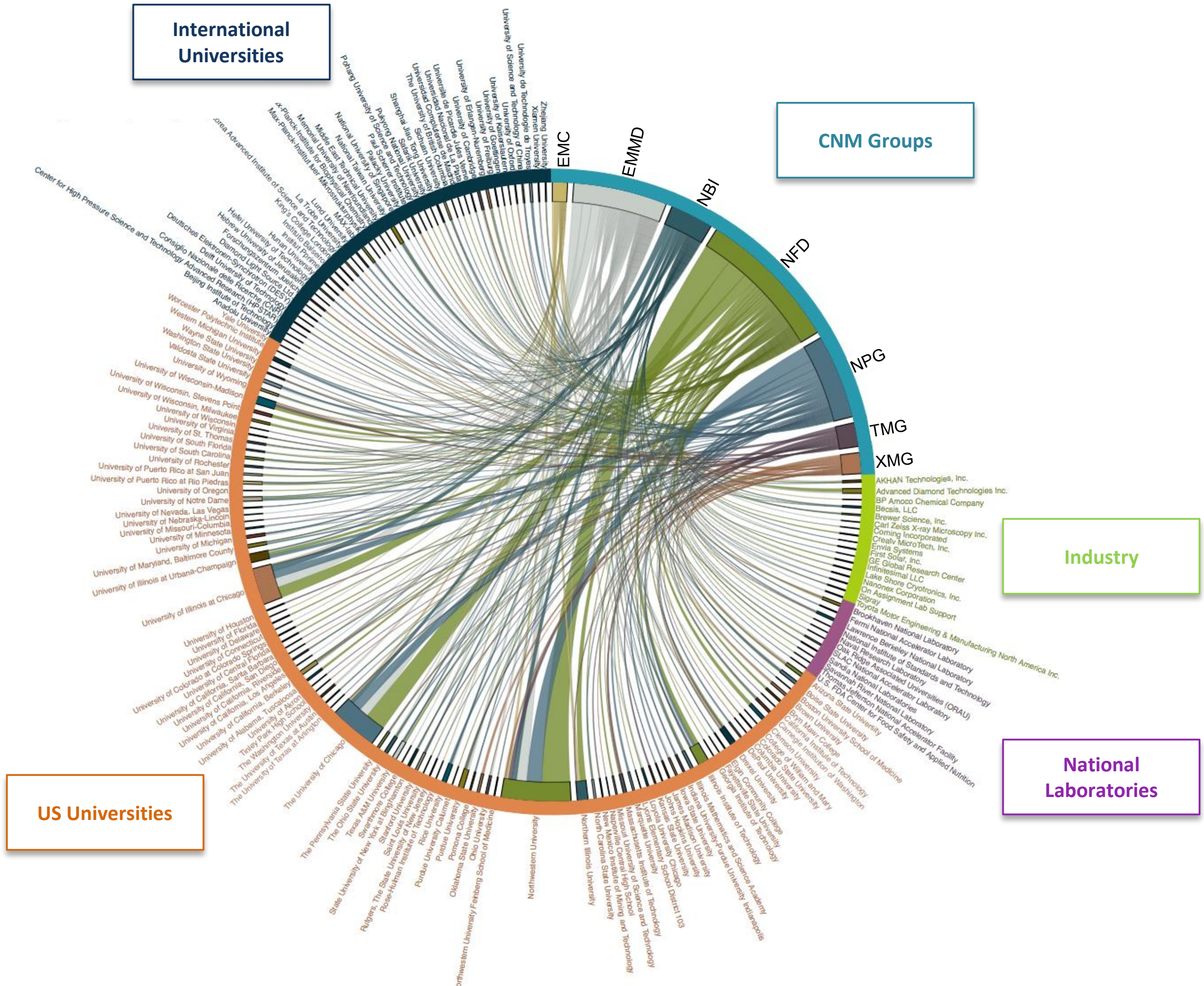


Figure 6-2. *User affiliation distributions (external to Argonne) during FY15 by CNM research group. EMMD stands for Electronic and Magnetic Materials and Devices that changed the name in FY2016 to QEM. NPG stands for Nanophotonics group and NBI for NanoBio Inetrface group. These two groups have merged in FY2016 into nPBS group; green = industrial; purple = other government; orange = U.S. academia; blue = international.*

Many of our capabilities and staff expertise are one-of-a-kind – people want to work at the CNM because it houses some of the most sophisticated and unique instrumentation together with scientists who can contribute intellectually to their endeavors. Our user partners are a diverse mix of university and government laboratory researchers—one of our goals going forward is to increase the participation of industrial users. To this end, we are coupled to a new organization, NanoDesignWorks, initiated at Argonne that focuses on technology commercialization particularly in the nanotechnology space. In keeping with the DOE's Office of Science mission of expanding knowledge and education, the CNM continues to provide internships to college and high school students. Quite often, the CNM's roughly twice a week tour of the facilities to visitors includes high school students. Very recently, we closed an agreement with United Scientific Supplies, a company that supplies science and laboratory kits to high schools in the US, to sell the "Nano-Fab-in-a-Box" kit that has been developed by CNM scientists.

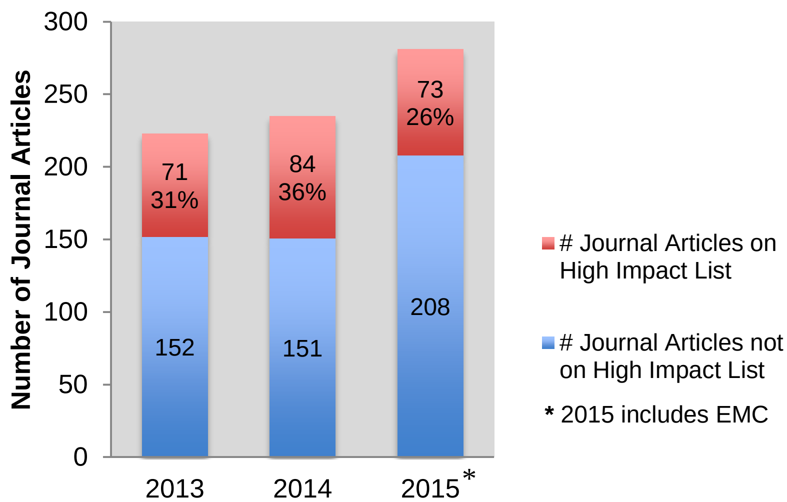


Figure 6-3. *Number and % of CNM journal articles in the NSRC high impact journal list versus those in other journals.*

The main outreach activities initiated by the CNM are focused towards enhancing our existing user community and to growing the user base in areas of the highest scientific impact. One example of the former is the annual users meeting held in conjunction with the APS to promote and enhance the latest research results within these co-located and complementary user communities. Over 500 registrants can select from new scientific workshop topics each year in addition to plenary sessions, poster sessions, short courses, an exhibitor venue, and various social events. All are welcome to attend the users meetings, including potential future users who wish to learn more about the CNM as they consider submitting user proposals. The joint CNM/APS Users Meetings are typically held during the second week of May. Activities targeted towards growing our user base in new areas of high scientific impact include promotion of our latest staff science results via professional scientific meetings, invited institutional talks, press releases, our public website and social media, as well as hosting other scientific workshops throughout the year. Examples of all of these events can be found on the CNM website at http://www.anl.gov/cnm/news.

7. Safety & Quality

The CNM has responsibility for environment, safety, health, and quality assurance (ESHQ) aspects of the facility's operations and, through policies and procedures, defines how responsibilities are delegated from the director through line managers to technically competent staff members supporting user research activities. The CNM program complements Argonne's laboratory-level safety program by incorporating methods, controls, and a work authorization approach tailored to the risk characteristics of a user facility and to the materials, instruments, and processes that constitute CNM operations. The specifics of the program will evolve in response to changing expectations, Argonne safety program evolution, and emerging information on hazards. Certified ES&H professionals help the CNM to better ensure research productivity and to ensure that the program efficiently implements applicable ESHQ standards and requirements.

Furthermore, the CNM continues to employ a precautionary approach where there is uncertainty about hazard potential of new chemicals, including nanomaterials. This concept guides the conduct of hazards analysis and specification of precautions when handling nanomaterials. The CNM has continued its efforts to contribute to a better understanding and management of ESH concerns associated with nanomaterials and ESH questions for nano-enabled products. Examples of relevant activities include the following:

- The CNM continues to participate in Argonne efforts to promote a consistent and effective approach to dealing with ESH concerns about nanomaterials, primarily through Argonne's Nanomaterial Safety Committee.
- The CNM is currently participating in the review of DOE Order 456.1, *The Safe Handling of Unbound Engineered Nanoparticles*. The CNM provides constructive criticism including comments reflecting the DOE Policy 456.1 requirement that the "best current knowledge is reflected in the identification and control of these potential hazards and impacts at their facilities."

Summary

In summary, we believe that the innovative science performed by CNM scientists shapes the user program while at the same time, innovative user science drives future scientific directions and capability development for the CNM. Sustaining this user-staff interaction, ensuring that the user base is distributed and diverse, making sure we remain relevant to user research needs in the future, and continuing to steward and shape the direction of the nanosciences remain our most important goals going forward.